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Theory of Collisionally Aided Radiative Excitation

in Three-level Systems: A New Interference Effect

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Collisionally Aided Radiative Excitation in a three-level atomic system is investigated using a stationary phase method for the case of large atom-field detunings and weak incident fields. It is found that collision-induced coherent phase interference effects can give rise to oscillatory structure in the total absorption cross section as a function of relative speed (energy). An example is given for a specific interatomic potential, indicating that experimental observation of such an effect is feasible.

The theory of saturation spectroscopy has led to an understanding of the physical processes that occur when two nearly resonant radiation fields are incident on a three-level atomic system. Collisional effects have been incorporated,[1] but such calculations have generally been limited to the impact region (|field detuning| << inverse collision time). Although calculations not restricted to the impact approximation have been carried out for two-level systems[2], there is, to our knowledge, only one calculation of collisionally aided radiative excitation (CARE, often referred to as "optical collision") in the non-impact limit for three-level systems.[3] This calculation was done using an effective two-level method and had serious restrictive conditions on the detunings.

In this Letter we report on a new type of quantum interference effect that occurs in three-level atomic systems subject to both collisions and off-resonant (|field detunings| >> inverse collision time) radiation fields. This effect results from an interference between contributions to the transition amplitudes from the various collision-induced crossings of the dressed atomic states[4] (see Fig. 1). When the crossings are well separated, the effect manifests itself as an oscillation in the total cross section as a function of active atom-perturber relative speed (energy). An analogous oscillatory feature was discussed by Rosenthal and Foley for charge-exchange inelastic collisions[5,6] in a He-He^+ system which is characterized by atom-ion interatomic potential curvessimilar to those in Fig. 1. However, there is an important difference between the two. In charge-exchange

inelastic collisions, the collision-induced crossings (or lack thereof) are completely determined by the atom-ion interatomic potentials. In CARE, on the other hand, the crossings are additionally dependent on the field detunings since they affect the dressed-atom level spacings.[7] Consequently, the nature and positions of the crossings may be altered in CARE (by varying the detunings) but not in charge-exchange inelastic collisions. This feature of CARE allows us to gain information on the diatomic systems which would be otherwise difficult to obtain.[8]

The system consists of a three-level active atom (levels labelled 1, 2, and 3) undergoing transitions from state 1 to state 3 simultaneously subjected to the collision of a structureless perturber and to two external fields of amplitudes E and E', frequencies ω and ω'. Fields E and E' drive only 1-2 (frequency ω_{21}) and 2-3 (frequency ω_{32}) transitions, respectively, their interaction being characterized by the detunings $\Delta = \omega - \omega_{21}$, $\Delta' = \omega' - \omega_{32}$ and the coupling strengths $\chi = \frac{\mu E}{2\hbar}$, $\chi' = \frac{\mu' E'}{2\hbar}$, where μ and μ' are the 1-2 and 2-3 dipole matrix elements. The collision is assumed to only shift the energies of the atomic levels without coupling them.

Figure 1 shows the system in a dressed-atom diabatic representation for some particular interatomic potentials and atom-field detunings; the atom-field dressed states are labelled I, II and III. The existence of crossings and their relative positions depend on the interatomic potential as well as the detunings.[9] Various crossing situations may ~~occur~~, leading to different features in the absorption cross section. We chose to focus our attention on the case when the detunings are large

($|\Delta|$, $|\Delta'|$ >> inverse collision time), when the field strengths are weak ($\chi << |\Delta|$, $\chi' << |\Delta'|$), and when there are three well-separated crossings, since an interesting interference phenomenon emerges under these conditions.

To better understand this interference effect, we use a time-dependent classical trajectory description of the collision event. For large detunings, states II and III can be excited directly from initial state I only at the crossing positions labelled by R_c and R_c'', respectively, in Fig. 1. In the time domain, the R_c and R_c'' crossings occur at corresponding times $\pm \tau_c$ and $\pm \tau_c''$ provided that the impact parameter is such that the distance of closest approach in the collision is less than the smaller of R_c and R_c''. Between their creation at times $\pm \tau_c$, $\pm \tau_c''$ and the crossing at time τ_c' (corresponding to internuclear distance R_c'), the amplitudes of states II and III evolve adiabatically with phases given by $\exp[-i\int_{\pm\tau_c}^{\tau_c'} W_{II}(R(t))dt]$ and $\exp[-i\int_{\pm\tau_c''}^{\tau_c'} W_{III}(R(t))dt]$, where W_{II}, and W_{III} are the collisionally modified energies shown in Fig. 1. At τ_c' transitions between states II and III are possible, and for times greater than τ_c' the amplitude of state III again evolves adiabatically.

Thus, there are four paths for excitation to state III, two from direct excitation I $\rightarrow$ III at $\pm \tau_c''$, the other two from stepwise excitation I $\rightarrow$ II $\rightarrow$ III at ($\pm \tau_c$, τ_c'), with each contributing to the probability amplitude. The cross term in the state III excitation probability contains six oscillatory terms varying as $\sin(\Delta\phi_i)$, $i = 1, 6$. Only one of these phases, $\Delta\phi_1 = \int_{\tau_c}^{\tau_c'} W_{II}(R(t))dt - \int_{\tau_c''}^{\tau_c'} W_{III}(R(t))dt$, is a slowly

varying function of the impact parameter b for the system under consideration. On integrating over b to obtain the excitation cross section, this term is the only one to survive. Since $\Delta\phi_1$ is essentially proportional to the time separation $\Delta\tau = (R_c' - R_c)/v$ (v is the relative atomic speed) between the inner and outer crossings, the total cross section oscillates as a function of $1/v$. These ideas are made more quantitative by the calculation given below.

The calculation is most conveniently carried out in the "bare" state picture. In the rotating wave approximation and weak field limit, the time dependent Schrödinger equation are solved by perturbation theory to yield the following expression for the state 3 amplitude $a_3(\infty)$, using as initial conditions $a_1(-\infty) = 1$, $a_2(-\infty) = a_3(-\infty) = 0$,

$$a_3(\infty) = -\chi\chi' \int_{-\infty}^{\infty} \exp\left\{-i\left[\Delta' t - \int_0^t V'(\tau)d\tau\right]\right\} \int_{-\infty}^{t} \exp\left\{-i\left[\Delta t_1 - \int_0^{t_1} V(\tau)d\tau\right]\right\} dt_1\, dt \qquad (1)$$

where V, V' are defined by $V(t) = V_2(t) - V_1(t)$, $V'(t) = V_3(t) - V_2(t)$ with $V_i(t)$ ($i = 1,2,3$) the collision-induced shift in energy of level i ($V_{1,2,3}(R) = W_{I,II,III}(R) + C_{1,2,3}$, where C_i's are constants). Owing to the condition |detunings| >> inverse collision time, we have neglected any Doppler phase shifts or level decays in Eq. (1).

Assuming that transitions occur only near the crossings, we use a stationary phase method to evaluate Eq. (1). For $R_c < R_c'' < R_c'$, the result is

$$|a_3(\infty)|^2 = A_s + A_D + A' \qquad (2)$$

with

$$A_s = (\chi\chi' 2\pi)^2 \left[1 + \delta \sin 2\phi\right] / (2\alpha\alpha') \qquad (3)$$

$$A_D = (\chi\chi'\pi)^2 (f^2 + g^2)\left[1 + \delta'' \sin 2(\phi'' - \delta\theta)\right] / (\alpha\alpha'') \qquad (4)$$

$$A' = -\delta \frac{(\chi\chi'\pi)^2}{\alpha}\left[\frac{2(f^2+g^2)}{\alpha'\alpha''}\right]^{1/2} \Big\{ \sin\left[\phi + \phi' - \phi'' + (\delta + \delta' - \delta'')\pi/4 + \delta\theta\right]$$
$$+ \sin\left[\phi + \phi' + \phi'' + (\delta + \delta' + \delta'')\pi/4 - \delta\theta\right]$$
$$+ \sin\left[\phi' + \phi'' - \phi + (\delta' + \delta'' - \delta)\pi/4 - \delta\theta\right]$$
$$+ \sin\left[\phi' - \phi'' - \phi + (\delta' - \delta'' - \delta)\pi/4 + \delta\theta\right] \Big\} \qquad (5)$$

where α, α' and α'' are one half of the absolute values of the time derivatives of $V(t)$, $V'(t)$, and $V''(t) = V(t) + V'(t)$, respectively, evaluated at τ_c, τ_c', and τ_c'', respectively, with the signs of these derivatives given by δ, δ', and δ'', respectively, f and g are the auxiliary functions of Fresnel's Integrals[10] with argument given by $|(2\alpha/\pi)^{1/2} (\tau_c'' - \tau_c)|$, and $\theta = \tan^{-1} g/f$.

The phases ϕ, ϕ', and ϕ'' can be written in a time-independent form; $\phi = (1/v) \int_b^{R_c} ((V(R) - \Delta)/(R^2 - b^2)^{1/2}) R dR$, where b is the impact parameter. Expressions for ϕ' and ϕ'' are of the same form with R_c, $V(R)$, and Δ replaced by R_c', $V'(R)$, Δ' and R_c'', $V''(R)$, Δ'', respectively. The only phase appearing in Eqs. (2-5) which is a slowly varying function of b is the one associated with the first term in A' since $\phi + \phi' - \phi''$ leads to integrals with limits independent of b (θ is slowly-varying in b). On

integrating Eqs. (2-5) over b to get the total cross section, the sine function in this term survives and oscillates as a function of 1/v.

Equations (2-5) are poor approximations near the impact parameter at which incoming and outgoing crossings coalesce, i.e. $b \approx R_c$. To obtain an accurate cross section, one must integrate the time dependent Schrödinger equation numerically near such impact parameters. However, a first approximation is achieved by assuming that all the sine terms in Eqs. (3-5) average to zero on integrating over b <u>except</u> the one varying as $\sin[\phi+\phi'-\phi''+(\delta+\delta'-\delta'')\pi/4+\delta\theta]$. In this term the phase is evaluated at $b = 0$ and the cross section is then approximated as $\sigma \simeq 2\pi\int_0^{R_c} |a_3(\infty)|^2\, b\,db$ to obtain

$$\sigma \simeq \frac{(\chi\chi'\pi)^2 R_c}{v^2\left|\left(\frac{dV}{dR}\right)_{R_c}\right|}\left\{\frac{4R_c'}{\left|\left(\frac{dV'}{dR}\right)_{R_c'}\right|}\ln\frac{R_c'+R_c}{R_c'-R_c}+\frac{2R_c''(f^2+g^2)}{\left|\left(\frac{dV''}{dR}\right)_{R_c''}\right|}\ln\frac{R_c''+R_c}{R_c''-R_c}\right.$$

$$\left.-2\delta\left[\frac{2R_c'R_c''(f^2+g^2)}{\left|\left(\frac{dV'}{dR}\right)_{R_c'}\left(\frac{dV''}{dR}\right)_{R_c''}\right|}\right]^{1/2}\ln\frac{R_c'+R_c''+2R_c}{R_c'+R_c''-2R_c}\cdot\sin(A/v+\phi_0)\right\} \tag{6}$$

where A is the area enclosed by the three crossing points and $\phi_0 = (\delta+\delta'-\delta'')\pi/4 + \delta\theta$ is a constant phase. A comparison of this cross section with the corresponding one obtained from computer solutions indicates that Eq. (6) is accurate to within 15%.

The calculation of cross section using Eq. (6) is remarkably simple. For given potential curves and detunings, one can graphically obtain

the slopes at the crossing points and the area A enclosed by the crossing points. Substitution of these values into Eq. (6) yields σ. The CARE cross section for the interatomic potential shown in Fig. 1 as a function of 1/v is shown in Fig. 2. The range of speed varies from 10^5cm sec^{-1} to 4×10^5 cm sec^{-1}. By varying the detunings, one can change A as well as the slopes at crossing points, and hence the frequency and amplitude of the oscillation in the total cross section. Although the example above is for a specific potential, we emphasize that the oscillatory feature occurs regardless of the form of the potential as long as three conditions are satisfied: first, there must be three crossings as in Fig. 1; second, the area enclosed by the crossings must be large enough to produce a phase change of the order π when the speed is varied in a convenient range; and third, the stepwise and the direct excitation contributions must be comparable. The first condition allows for a phase factor that is nearly b-independent, and the second and the third conditions determine the frequency and amplitude of the oscillatory term.

For the potential and detunings shown in Fig. 1, the excitation cross sections are of the order of $(10^{-34}\ I\ I')$cm^2 with I, I' the power density in W/cm^2. Thus the effect should be observable with moderate laser power. The experiment must be performed using crossed atomic beams or a beam interacting with a gas sample. The beam-gas sample method works only if the active-atom-perturber relative velocity is approximately equal to the beam velocity.

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Figure Captions

Figure 1

Collisionally-modified dressed-state energies W_I, W_{II}, W_{III} of a three-level active atom and applied fields as a function of internuclear separation, R, in the weak field limit. The energies $\hbar|\Delta|$ and $\hbar|\Delta'|$ set the energy scale. $\Delta = -8\times10^{13}$ sec^{-1}, $\Delta' = -3\times10^{13}$ sec^{-1}.

Figure 2

Total excitation cross section as a function of inverse relative speed v for a potential shown in Fig. 1 with $\chi = \chi' = 10^{11}$ sec^{-1}, $\Delta = -8\times10^{13}$ sec^{-1}, and $\Delta' = -3\times10^{13}$ sec^{-1}. The curve rises as $(1/v)^2$. As the speed varies from 10^5 cm sec^{-1} to 4×10^5 cm sec^{-1}, equally spaced peaks are clearly seen. In the inset, (total cross section x v^2) as a function of 1/v is shown.

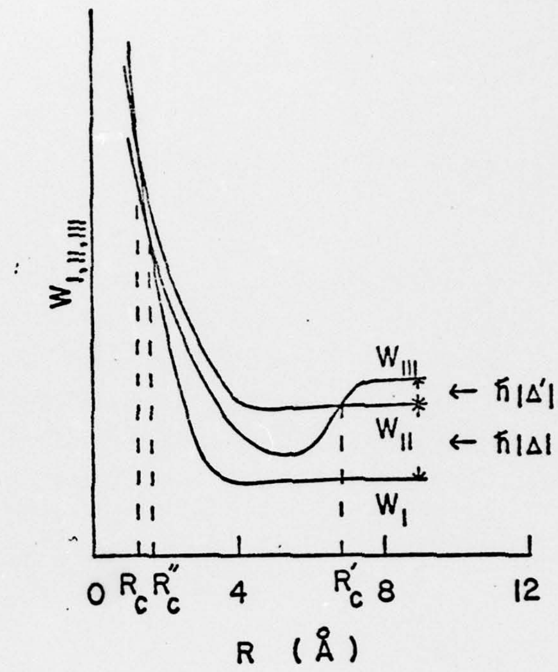

Fig. 1

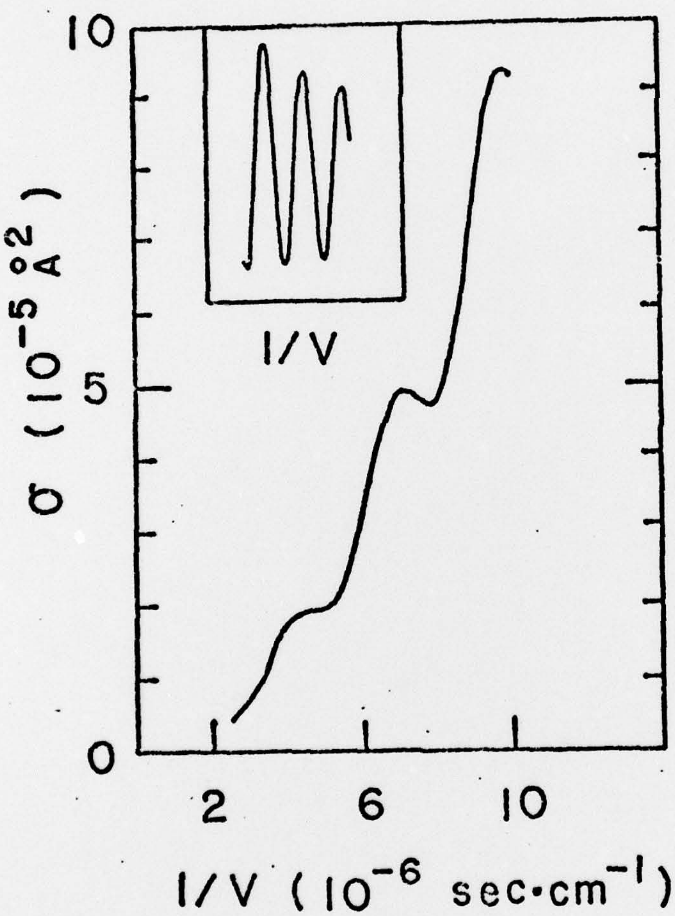

Fig. 2